Size and strain effects in the E_1-like optical transitions of InAs/InP self-assembled quantum dot structures

J. A. Prieto[a] and G. Armelles
Instituto de Microelectrónica de Madrid (C.N.M., C.S.I.C.), Isaac Newton 8, 28760–Tres Cantos (Madrid), Spain

J. Groenen and R. Carles
Laboratoire Physique des Solides, Université P. Sabatier, 118 route de Narbonne, 31062 Toulouse-Cédex, France

(Received 17 June 1998; accepted for publication 25 October 1998)

The optical transitions of uncapped and capped InAs/InP self-assembled quantum dot structures in the energy range of the bulk InAs E_1 transition are studied using modulation spectroscopy and Raman scattering. Islands and wetting layer exhibit one and two features, respectively. The deformation potential theory and the single band effective mass approximation account for the island-related feature successfully in a wide island-height range (3–7 nm). These models also explain the existence of the highest energy wetting layer related feature, but not of the lowest energy one. © *1999 American Institute of Physics.* [S0003-6951(99)00101-1]

Despite the strong effort devoted to the study of the optical properties of semiconductor quantum dots (QDs), little attention has been paid to their above band-gap transitions. However, the study of these transitions is important in view of some recent theoretical and experimental results. An example of these results is the prediction of large couplings between the bulk Γ, L, and X states at nanometer size.[1] The understanding of the phenomenology related to the E_1- or E_2-like (L or X derived) transitions may provide a better comprehension of such interband mixings and, therefore, of the electronic structure of semiconductor QDs. Another example is the facility in estimating the strain in self-assembled QDs by means of the combination of the analysis of the E_1-like transition and Raman scattering measurements.[2,3] This facility rests on the fact that, due to the large effective masses of carriers, the effect of quantum confinement on the bulk L states is negligible for big enough islands, as compared with that on the bulk Γ states. However, quantum confinement may play an important role in smaller islands. In the present letter we report on the signatures of the InAs/InP self-assembled QD system about the bulk InAs E_1 transition. These signatures are detected and assigned to the islands or to the wetting layer (WL) using modulation spectroscopy and Raman scattering. The energy of the island-related E_1-like transition is simulated taking into account both quantum confinement and strain within the deformation potential theory and the single band effective mass approximation. The validity of these models is thus examined.

Three different sets of (001)-oriented samples are studied here. The first set was grown under conditions which favor island formation, consisting of two uncapped specimens with InAs deposits of 3.5 and 5 monolayers (ML), respectively. Atomic-force microscopy studies reveal a similar mean island height, 5.8 nm, in both of them. The island coverage increases with the InAs deposition, as should be expected. Island formation was also favored in the growth of the second set, formed by three capped specimens with InAs deposits of 1.8, 2.5, and 4 ML, respectively; the InP cap layer thickness is 25 nm. Also in this case, the island coverage augments with the InAs deposition. The 1.8 ML sample has a bimodal island-height distribution, with values of the mean island height of 3.5 and 7.4 nm and a dispersion much larger for the smallest than for the biggest islands. On the contrary, the 2.5 and 4 ML samples do have a single island-height distribution and values of the mean island height of 3.3 and 3.6 nm, respectively. The structural aspects of these three samples are reported in detail elsewhere.[4] Finally, the third set comprises only a 2-ML-thick InAs quantum well (QW) sample, grown under conditions which avoid island formation. This QW was covered, like the islands of the capped specimens, with a 25-nm-wide InP cap layer. The structural characterization[5] reveals the existence of a strong corrugation at the InAs/InP interface or very small thin QDs, but not of well-developed QDs.

Figure 1 shows the photoreflectance (PR) spectra of the uncapped samples taken at 80 K. The features located between 3.0 and 3.4 eV correspond to the E_1 and $E_1+\Delta_1$ transitions of the underlying InP, whereas the one around 2.65 eV is associated with the InAs deposit. As the increase of the signal-to-noise ratio indicates, the intensity of this feature rises, like the island coverage, with the InAs deposition. In addition, its energy is, in the same way as the mean island height, similar in both samples. This feature is therefore related to the uncapped islands.

Several InAs-related features are observable in the PR and electroreflectance (ER) spectra of the capped samples recorded at 80 K (Fig. 2). We attribute the ones near 2.7 eV to the buried islands with regard to: First, the feature of the 4 ML sample is situated at lower energy than that of the 2.5 ML sample, as should be expected from the larger value of the mean island height corresponding to the former one; and second, the 1.8 ML sample exhibits two features in accor-

[a]Electronic mail: josea@imm.cnm.csic.es

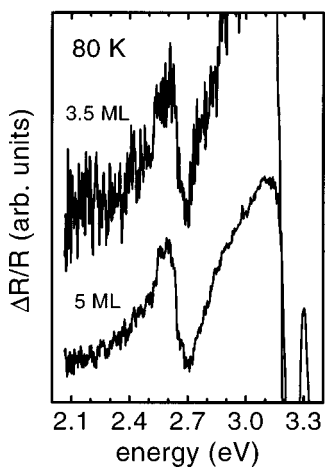

FIG. 1. 80 K PR spectra of two uncapped InAs/InP QD structures in the spectral range of the bulk InAs E_1 transition. The nominal InAs deposits are 3.5 and 5 ML.

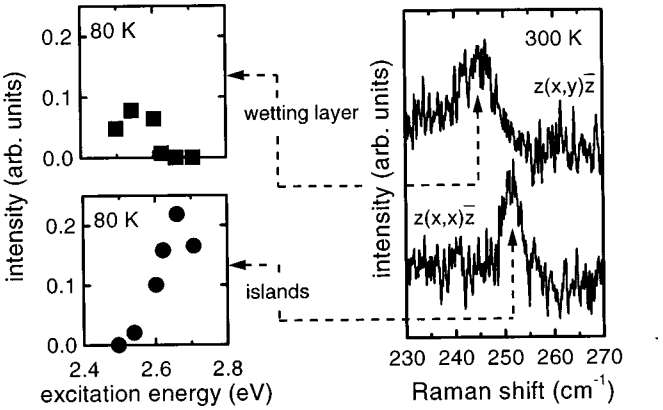

FIG. 3. Right: 300 K Raman spectra of the capped InAs/InP QD structure with nominal InAs deposit of 1.8 ML recorded in the crossed $[z(x,y)\bar{z}]$ and parallel $[z(x,x)\bar{z}]$ light polarization configurations (x, y, and z denote the [100], [010], and [001] directions, respectively). Both of them display InAs-related LO(Γ)-like phonons, the former coming from the wetting layer and the latter one from the islands. Left: 80 K resonant profiles corresponding to the wetting layer (above ■) and the islands (below, ●).

dance with its bimodal island-height distribution. Moreover, Raman scattering by the longitudinal optical (LO) phonon related to the buried islands[5] displays a resonance close to 2.7 eV (Fig. 3). Another Raman scattering resonance, around 2.5 eV, appears in the LO phonon of the WL[5] (Fig. 3). This feature is perceptible in the ER spectrum of the 4 ML sample. Also, it could be hidden by the lowest energy island-related one in the 1.8 ML sample and by the Franz–Keldish oscillations coming from the InP matrix in the 2.5 ML sample. There exists another feature in all these spectra, above 2.9 eV, that must be also connected with the WL: The intensity ratio between this and the island-related features diminishes with the InAs deposition as a result of the decrease of the WL volume related to the increase of the island coverage. The feature located above 2.9 eV appears also in the ER spectrum of the QW sample (Fig. 2). In addition, a signature that resembles those coming from the QDs can be observed at 2.8 eV. Its broadening parameter is, however, much smaller, suggesting an origin probably related to the strong interface corrugation or the very small thin islands existing in this sample.

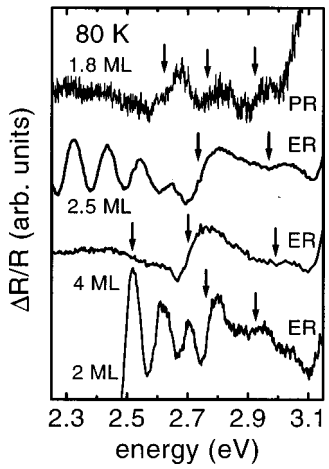

FIG. 2. 80 K PR and ER spectra of capped InAs/InP QD structures in the spectral range of the bulk InAs E_1 transition. The nominal InAs deposits are 1.8, 2.5, and 4 ML. Also, the ER spectrum of a 2-ML-thick InAs QW sample is presented for comparison. The InAs-related features are indicated by arrows.

In view of the energy range considered in this work, it is tempting to assign the InAs-related features to the E_1-like transitions of the islands and the WL. The energies of these transitions have been calculated from the bulk parameters taking into account both quantum confinement and strain. Due to the very small aspect ratio (height/base) of the islands and the large effective masses of the carriers of the bulk L states, quantum confinement is significant only along the growth direction. In consequence, the islands can be modeled as a quantum well of width equal to the mean island height and depth determined by the strain-dependent band alignment. As shown by recent theoretical calculations,[6,7] the mean values of the strain components of an island satisfy the following relationships: $\epsilon_{xx} \approx \epsilon_{yy}$, $\epsilon_{zz}/\epsilon_{xx} = (-2C_{12}/C_{11}) \times \alpha$, $\epsilon_{xy} \approx \epsilon_{xz} \approx \epsilon_{yz} \approx 0$, where C_{11} and C_{12} are bulk elastic stiffness constants and α is a numerical parameter close to one. Assuming $\alpha = 1$, these components were obtained for our islands from the frequency of the island-related LO phonon.[8] Then, the band alignment of the strained L-like states was determined from the modifications caused by the strain in the energies of the conduction band edge and the E_1 transition of bulk material.[9] We used the deformation potentials of GaAs (Ref. 9) as a first approach and the valence band offset for the Γ-like states and the relative positions of the L band edges of InAs and InP reported in Refs. 10 and 11, respectively. The result of these calculations is depicted in Fig. 4; the case of InAs lattice matched to InP is also shown for the sake of completeness. The effect of quantum confinement on the energy levels of holes and electrons was evaluated within the single band effective mass approximation,[12] using the [001] projections of the L effective masses of GaAs.[12–14] In the case of the uncapped islands, the vacuum barrier energies were taken from Ref. 15. Figure 5 displays the experimental energies of the island-related features, which were obtained fitting to the first derivative of a lorentzian functional form,[16] as well as the calculated energies. We should mention that moderate changes in α do not alter significantly these energies. Good agree-

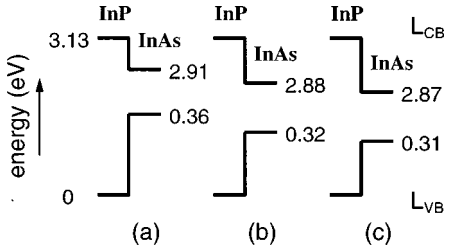

FIG. 4. Band alignments at the L point of the Brillouin zone obtained within the deformation potential theory for (a) InAs lattice matched to InP, (b) InAs islands deeply buried in InP, and (c) InAs islands on top of InP. The relationships between the mean strain components $\epsilon_{xx}=\epsilon_{yy}$, $\epsilon_{zz}/\epsilon_{xx}=-2C_{12}/C_{11}$, $\epsilon_{xy}=\epsilon_{xz}=\epsilon_{yz}=0$ have been assumed, with $\epsilon_{xx}=-3.2\times10^{-2}$ in (a), -2.9×10^{-2} in (b), and -2.5×10^{-2} in (c).

ment exists between experimental and calculations, with the exception of the highest energy feature of the 1.8 ML capped sample. This sample has a rather large dispersion in the height of the smallest islands. Since that feature is the convolution of the transitions coming from islands with very different heights, the value of the mean island height is not as well defined as for the biggest islands of this sample and for the islands of the other samples. Our simple calculations account for the island-related feature successfully in a wide island-height range. In consequence, neither band coupling nor interband mixing should be taken into consideration to describe the island-related E_1-like transition for islands higher than 3 nm. These calculations also explain the origin of the highest energy WL-related transition, but do not yield the correct value of its energy, as might be expected from the very small WL thickness. However, the existence of the lowest energy WL-related transition is completely inexplicable within such a framework.

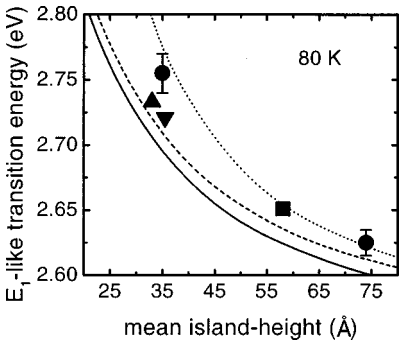

FIG. 5. E_1-like transition energy vs the mean island height for InAs islands deeply buried in InP (dashed line) and InAs islands on top of InP (dotted line) estimated at 80 K. The case of an InAs QW strained to InP is also presented (solid line). The calculations have been carried out, from the band alignments depicted in Fig. 4, within the single band effective mass approximation. The experimental energies are shown by symbols: uncapped islands, 3.5 and 5 ML samples (■); capped islands, 1.8 ML sample (●), 2.5 ML sample (▲), and 4 ML sample (▼). Error bars are displayed when bigger than symbols.

In summary, three transitions, one coming from the islands and two from the WL, have been observed around the bulk InAs E_1 transitions in the InAs/InP self-assembled QD system. The deformation potential theory and the single band effective mass approximation describe successfully the island-related E_1-like transition for values of island height larger than 3 nm, where the contributions of quantum confinement and strain have been found to be similar. Such models also explain the origin of the highest energy WL-related transition; however, more complex theoretical treatments are needed to justify the existence of the lowest energy one.

This work has been supported by the Spanish CICYT under Grant No. MAT95-0966-C02-01 and by the French–Spanish program 96/240 B.

[1] H. Fu and A. Zunger, Phys. Rev. Lett. **80**, 5397 (1998).
[2] P. D. Persans, P. W. Deelman, K. L. Stokes, L. J. Schowalter, A. Byrne, and T. Thundat, Appl. Phys. Lett. **70**, 472 (1997).
[3] J. A. Prieto, G. Armelles, T. Utzmeier, F. Briones, J. C. Ferrer, F. Peiró, A. Cornet, and J. R. Morante, Phys. Rev. Lett. **80**, 1094 (1998).
[4] A. Ponchet, A. Le Corre, H. L'Haridon, B. Lambert, and S. Salaün, Appl. Phys. Lett. **67**, 1850 (1995).
[5] J. Groenen, A. Mlayah, R. Carles, A. Ponchet, A. LeCorre, and S. Salaün, Appl. Phys. Lett. **69**, 943 (1996).
[6] W. Yu and A. Madhukar, Phys. Rev. Lett. **79**, 905 (1997).
[7] C. Priester (private communication).
[8] F. Cerdeira, C. J. Buchenauer, F. H. Pollak, and M. Cardona, Phys. Rev. B **5**, 580 (1972).
[9] F. H. Pollak, in *Strained-Layer Superlattices: Physics, Semiconductors and Semimetals*, edited by T. P. Pearsall (Academic, San Diego, 1990), Vol. 32, Chap. 2.
[10] R. Houdré, J. F. Carlin, A. Rudra, J. Ling, and M. Ilegems, Superlattices Microstruct. **13**, 67 (1993).
[11] Landolt-Börnstein, *Numerical Data and Functional Relationships in Science and Technology, New Series*, edited by O. Madelung (Springer, Berlin, 1987), Vol. 22a.
[12] E. E. Mendez, L. L. Chang, G. Landgren, R. Ludeke, L. Esaki, and F. H. Pollak, Phys. Rev. Lett. **46**, 1230 (1981).
[13] D. E. Aspnes, Phys. Rev. B **14**, 5331 (1976).
[14] S. Adachi, J. Appl. Phys. **58**, R1 (1985).
[15] J. M. Sallese, J. F. Carlin, M. Gailhanou, and M. Ilegems, Appl. Phys. Lett. **71**, 2331 (1997).
[16] W. M. Theis, G. D. Sanders, C. E. Leak, K. K. Bajaj, and A. Morkoç, Phys. Rev. B **37**, 3042 (1988).